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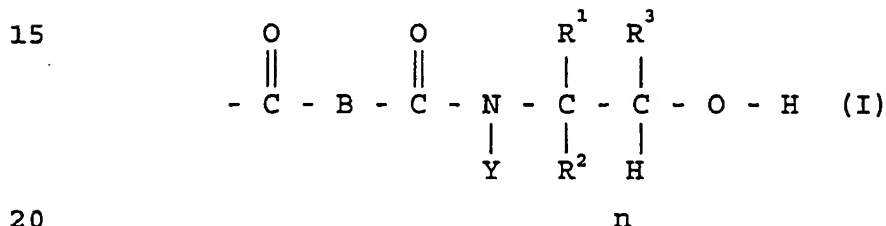
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CONDENSATION POLYMER CONTAINING HYDROXYALKYLAMIDE
GROUPS

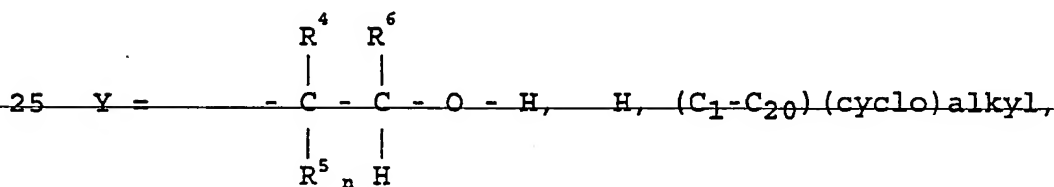
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The invention relates to a linear or branched condensation polymer containing ester groups and at least one amide group in the backbone, having at least one hydroxyalkylamide endgroup and having a weight average molecular mass of ≥ 800 g/mol.

Preferably, the polymer contains at least two groups according to formula (I)



in which



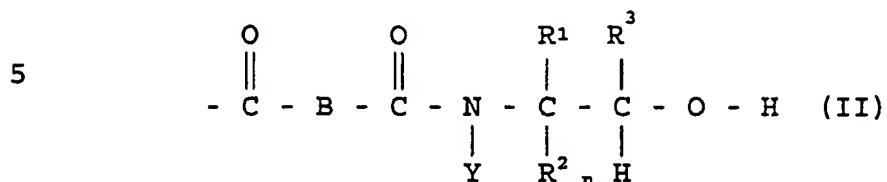
or (C₆-C₁₀) aryl,

30 B = (C₂-C₂₄), optionally substituted, aryl or (cyclo)alkyl aliphatic diradical,

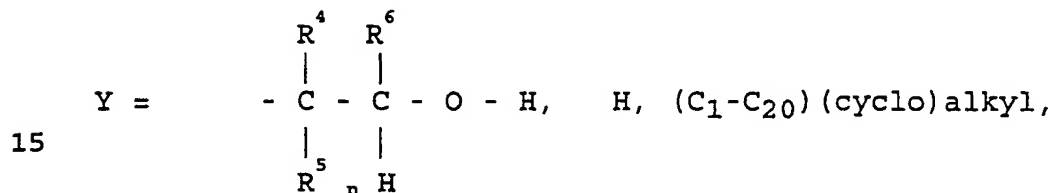
R¹, R², R³, R⁴, R⁵ and R⁶ may, independently of one another, be the same or different, H, (C₆-C₁₀) aryl or (C₁-C₈) (cyclo)alkyl radical and

35 n = 1-4.

More preferably the polymer contains at least two groups according to formula (II):



10 in which

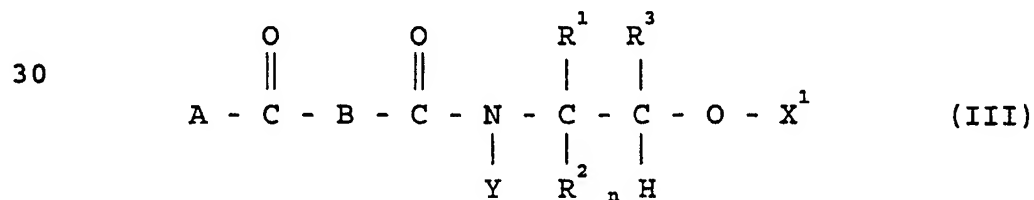


or (C₆-C₁₀) aryl,

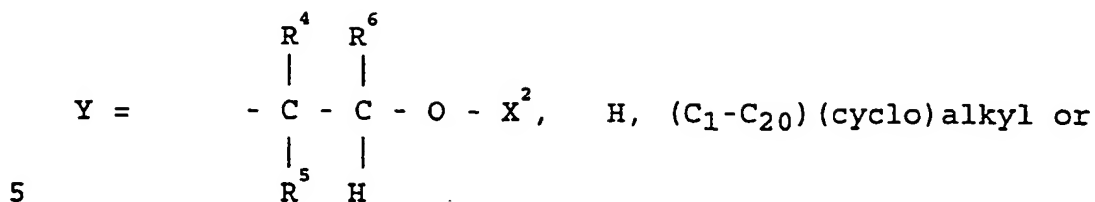
20 B = (C₂-C₂₄), optionally substituted, aryl or (cyclo)alkyl aliphatic diradical, and

R¹, R², R³, R⁴, R⁵ and R⁶ may, independently of one another, be the same or different, H, (C₆-C₁₀) aryl or (C₁-C₈) (cyclo)alkyl radical.

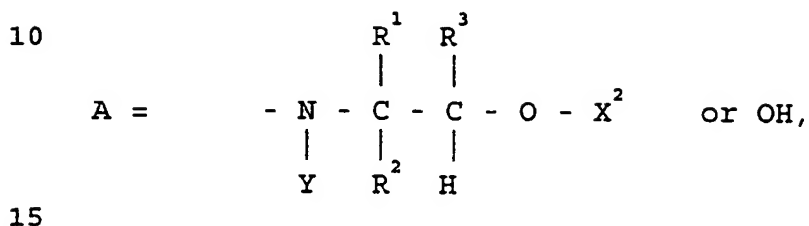
25 According to a further preferred embodiment, the polymer containing hydroxyalkylamide groups is a polymer according to formula (III):



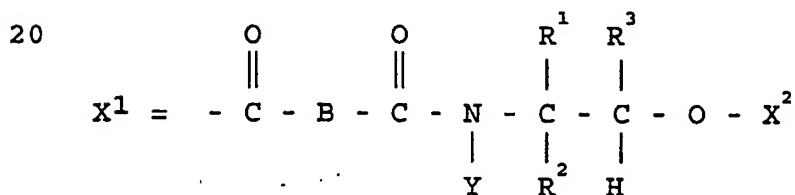
35 in which:



(C₆-C₁₀) aryl



B = (C₂-C₂₄), optionally substituted, aryl or
(cyclo)alkyl aliphatic diradical,



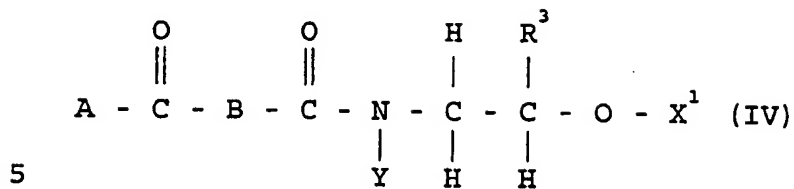
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X² = H or X¹ and

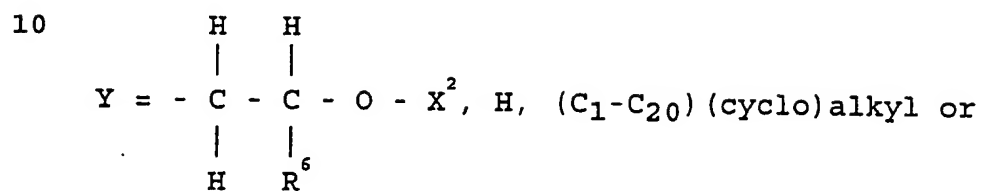
R¹, R², R³, R⁴, R⁵ and R⁶ may, independently of one
another, be the same or different, H, (C₆-C₁₀) aryl or
30 (C₁-C₈) (cyclo)alkyl radical or CH₂-OX².

In formulas (I), (II) and (III) R groups
may together or with neighbouring carbon atoms form
part of a cycloalkyl group.

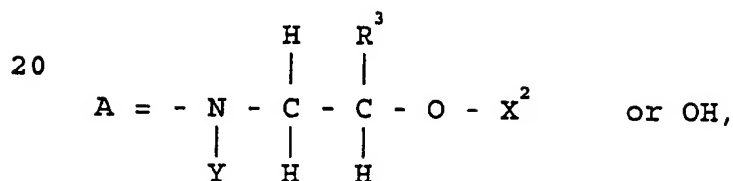
According to another preferred embodiment
35 of the invention, the polymer containing β-
hydroxyalkylamide groups is a polymer according to
formula (IV):



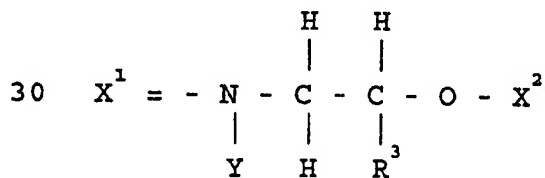
in which:



(C₆-C₁₀)aryl,



25 B = (C₂-C₂₀), optionally substituted, aryl or
(cyclo)alkyl aliphatic diradical,



35 X² = H or X¹,

R³ = H or (C₆-C₁₀) aryl or (C₁-C₈)alkyl radical and

R⁶ = H or (C₆-C₁₀) aryl or (C₁-C₈)alkyl radical.

The weight average molecular mass of the polymer according to the invention is generally between

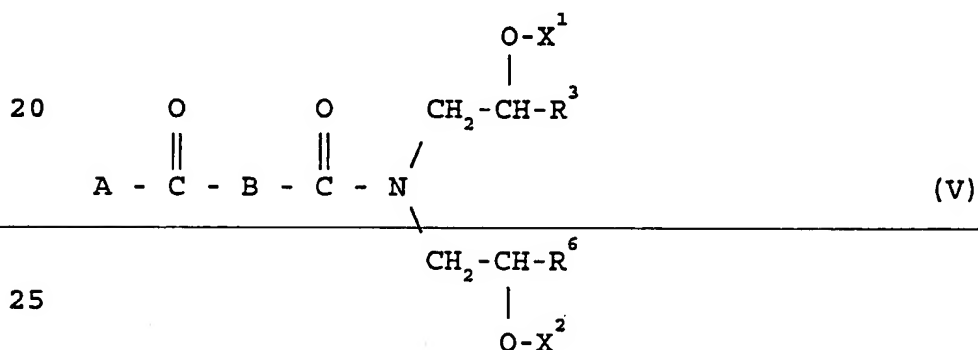
800 and 50,000, and preferably between 1000 g/mol and 25,000 g/mol.

The number average molecular mass is generally between 600 and 10,000 and preferably between 700 and 4000.

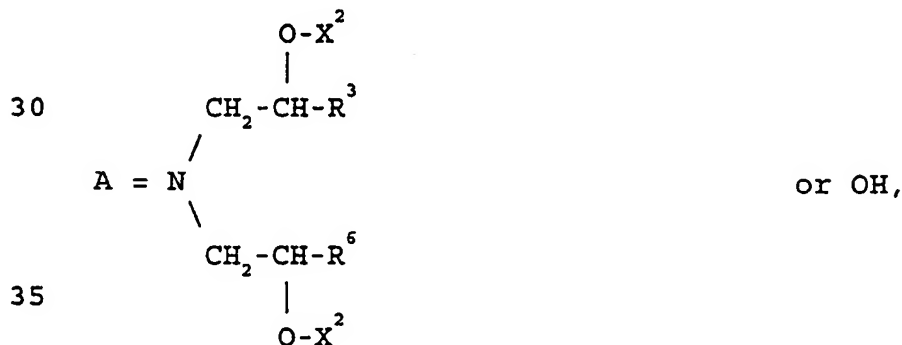
The hydroxyalkylamide functionality is generally between 2 and 250 and preferably between 5 and 50.

Functionality is the average number of reactive groups of the specific type per molecule in the polymer composition.

According to another preferred embodiment of the invention the polymer's hydroxyalkylamide functionality of the polymer is ≥ 5 and the polymer containing β -hydroxyalkylamide groups is a polymer represented by formula (V):

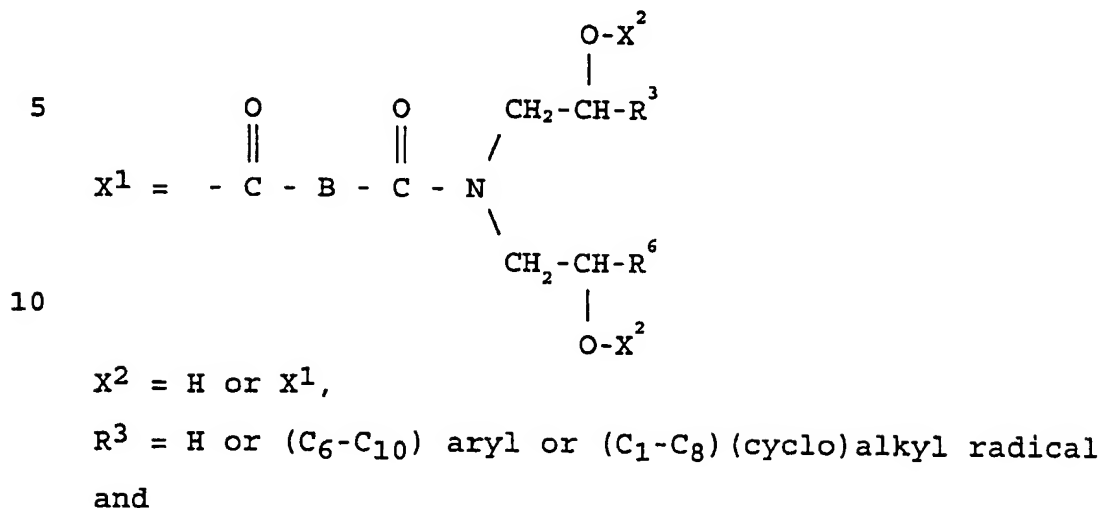


in which:



B = (C₂-C₁₂), optionally substituted, aryl or

(cyclo)alkyl aliphatic diradical,



$\text{R}^6 = \text{H or (C}_6\text{-C}_{10}) \text{ aryl or (C}_1\text{-C}_8) \text{ (cyclo)alkyl radical}$

Preferably R^3 and R^6 are $(\text{C}_1\text{-C}_4)$ alkyl.

According to another preferred embodiment of the invention R^3 and R^6 are methyl or ethyl.

B may be saturated or unsaturated.

B may be substituted with for example a $(\text{C}_1\text{-C}_{26})$ alkyl group, which may be saturated or unsaturated; preferably C_1 is used.

B may be for example a (methyl-)1,2-ethylene, (methyl-)1,2-ethylidene, 1,3-propylene, (methyl-)1,2-cyclohexyl, (methyl-)1,2-phenylene, 1,3-phenylene, 1,4-phenylene, 2,3-norbornyl, 2,3-norbornen-5-yl and/or (methyl-)1,2 cyclohex-4-enyl radical.

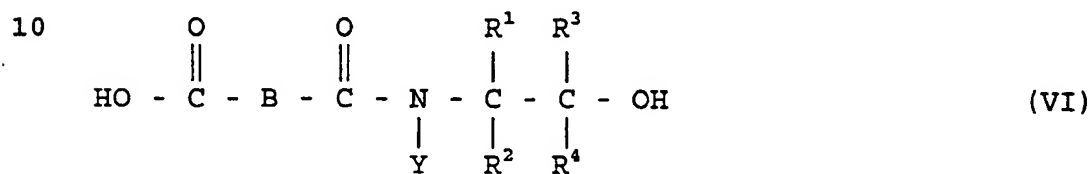
Depending on the starting monomers chosen, the variables B, R^1 , R^2 , R^3 , R^4 , R^5 and R^6 in the molecule or mixture of molecules can be selected to be the same or different per variable.

The polymer composition according to the invention is generally a composition comprising higher and lower oligomers, which usually contains less than 50 wt.%, preferably less than 30 wt.%, of oligomers

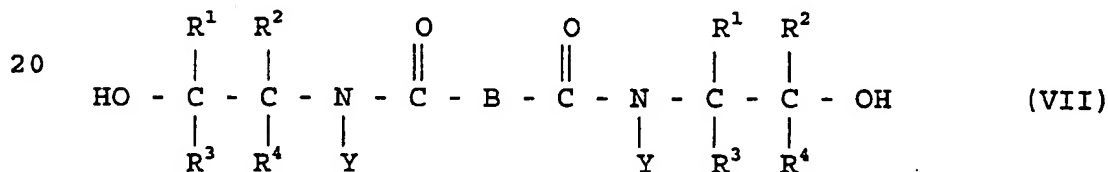
having a molecular weight smaller than 600.

The polyesteramide according to the invention can for example be obtained through polycondensation of mono- and/or bis-hydroxyalkylamides
5 of bivalent carboxylic acids.

The monohydroxyalkylamide of a bivalent carboxylic acid generally has the formula (VI):



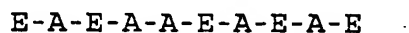
and the bishydroxyalkylamide of a bivalent carboxylic acid generally can be represented by formula (VII):



wherein

R^1 , R^2 , R^3 and R^4 may, independently of one another, be the same or different, H, (C_6 - C_{10}) aryl or (C_1 - C_8) (cyclo)alkyl radical.

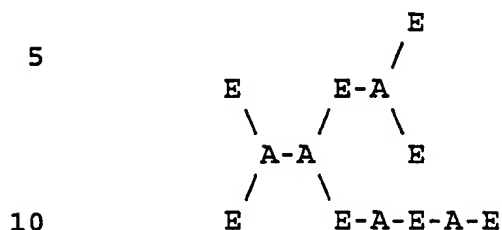
Consequently a linear polymer according to the invention generally comprises the amide and the ester groups alternating along the chain as follows:



wherein a diamide is coupled with alternating ester (E) - amide (A) groups.

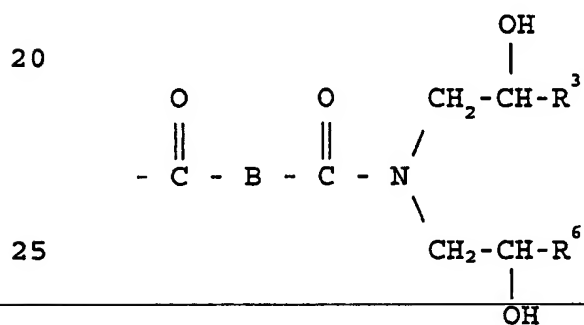
A branched polymer according to the invention generally comprises the amide and the ester

groups alternating along the main and side chains as follows:

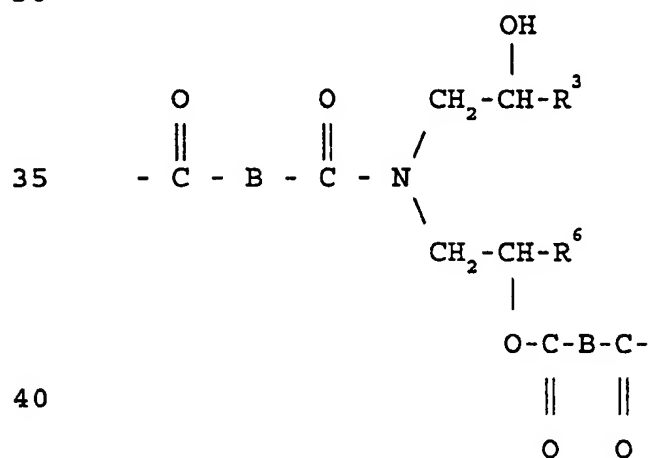


wherein a diamide is coupled with alternating ester (E) - amide (A) groups.

In the branched polymers according to the invention (β)-hydroxyalkylamide groups can be present both as an endgroup



and as a pendant side chain group



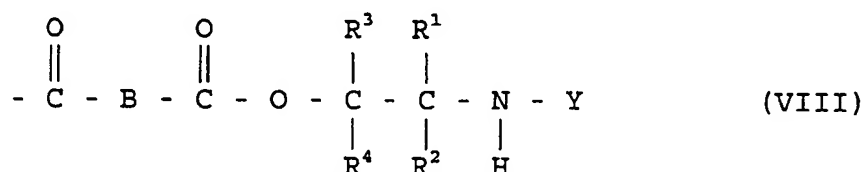
Generally, the molar amount of amide bounds

in the chain is higher than the amount of ester bounds.

The polymer according to the invention comprises at least 60% by weight of the products represented by the formulas (III)-(V).

5 Due to side reactions during the preparation of the polymer it is possible that the composition according to the invention comprises also for example secondary amine groups having the formula (VIII):

10



15

wherein:

R^1 , R^2 , R^3 and R^4 may, independently of one another, be the same or different, H, $(\text{C}_6\text{-C}_{10})$ aryl or $(\text{C}_1\text{-C}_8)$ (cyclo)alkyl radical.

20

The polymer according to the invention can, also be obtained in a one-step procedure by reacting a cyclic anhydride and an alkanolamine, at a temperature between for example about 20°C and about 100°C, to form

25 a hydroxyalkylamide, after which, at a temperature between, for example, 120°C and 250°C, a polyesteramide is obtained through polycondensation with water being removed through distillation.

The reaction can take place without a solvent, but also in water or in an organic solvent.

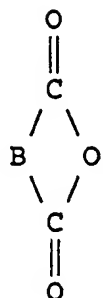
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The removal of water through distillation can take place at a pressure higher than 1 bar, in a vacuum or azeotropically.

Preferably, the cyclic anhydride is an anhydride according to formula (IX):

35

5



(IX)

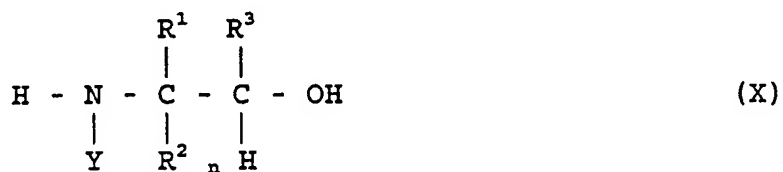
10

in which B has the meaning specified above.

Examples of suitable cyclic anhydrides include phthalic anhydride, tetrahydrophthalic anhydride, naphthalenic dicarboxylic anhydride, hexahydrophthalic anhydride, 5-norbornene-2,3-dicarboxylic anhydride, norbornene-2,3-dicarboxylic anhydride, naphthalenic dicarboxylic anhydride, 2-dodecene-1-yl-succinic anhydride, maleic anhydride, (methyl)succinic anhydride, glutaric anhydride, 4-methylphthalic anhydride, 4-methylhexahydrophthalic anhydride, 4-methyltetrahydrophthalic anhydride and the maleinised alkylester of an unsaturated fatty acid.

Preferably the alkanol is an alkanolamine according to formula (X):

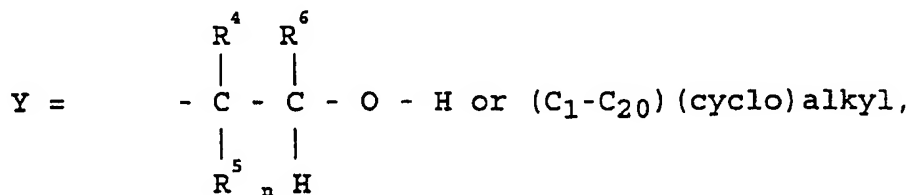
25



30

in which:

35



40

R¹, R², R³, R⁴, R⁵ and R⁶ may, independently of one another, be the same or different, H, (C₆-C₁₀) aryl or (C₁-C₈) (cyclo)alkyl radical or CH₂OH and n=1-4.

More preferably n=1. The alkanolamine may
5 be a monoalkanolamine, a dialkanolamine, a trialkanolamine or a mixture hereof.

If monoalkanolamines are used in one of the possible polymer syntheses, linear polymers with a functionality of 2 can be obtained. Depending on the
10 application desired, a linear or an entirely or partly branched polymer can be chosen, in which case the degree of branching can be set via the alkanolamines chosen.

If a highly branched structure with a high
15 functionality is desired, di- or trialkanolamines are used as the starting compound.

Examples of suitable mono- β -alkanolamines include ethanolamine, 1-(m)ethyl ethanolamine, n-butyl
20 ethanolamine, 1-(m)ethyl isopropanolamine, ~~isobutanolamine, β -cyclohexanolamine, n-butyl~~
isopropanolamine and n-propanolamine.

Examples of suitable di- β -alkanolamines are 3-amino-1,2-propanediol, 2-amino-1,3-propanediol
25 diisobutanolamine (bis-2-hydroxy-1-butyl)amine), di- β -cyclohexanolamine and diisopropanolamine (bis-2-hydroxy-1-propyl)amine).

A suitable trialkanolamine is, for example, tris(hydroxymethyl)aminomethane.

30 Preferably a β -alkyl-substituted β -hydroxyalkylamide is used. Examples are (di)isopropanolamine, cyclohexyl isopropanolamine, 1-(m)ethyl isopropanolamine, (di)isobutanolamine, di- β -

cyclohexanolamine and/or n-butyl isopropanolamine.

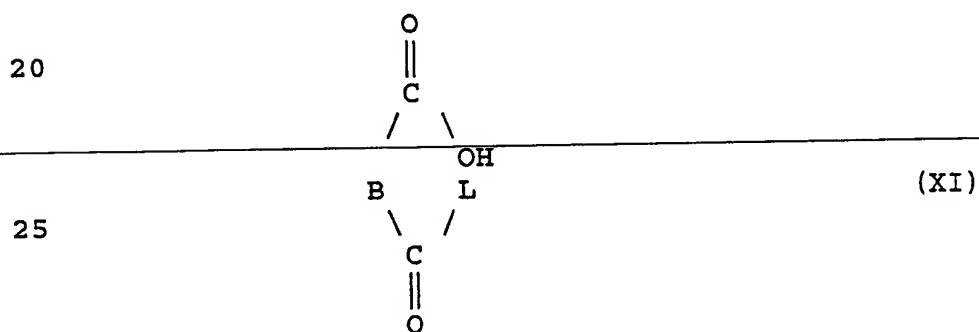
This results in polymer compositions with improved resistance to hydrolysis.

Most preferable are diisopropanolamine and
5 diisobutanolamine.

The anhydride : alkanolamine equivalent ratio is generally between 1.0:1.0 and 1.0:1.8. Preferably, this ratio is between 1:1.05 and 1:1.5.

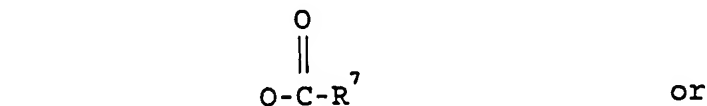
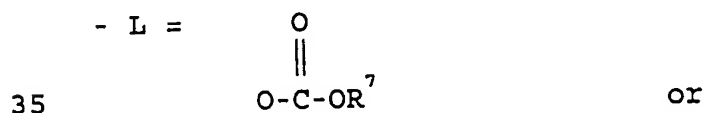
The compound according to the invention can
10 also be obtained via a reaction between an alkanolamine, as for example described above, and a compound containing one acid group and one activated acid group, after which a polyesteramide is obtained through polycondensation.

15 The compound containing an acid group and an activated acid group is preferably a compound according to formula (XI):



30 in which

- B has the meaning specified above and



40

OR⁷

or

SR⁷

or

5

Cl

in which R⁷ is a (C₁-C₁₂) branched or linear alkyl group.

Examples of suitable compounds containing one acid group and one activated acid group are alkyl esters, such as, for example, mono(m)ethyl adipate and mono(m)ethyl sebacate, anhydrides and thioesters.

The compound according to the invention can also be obtained via a reaction between a cyclic anhydride, as for example described above, and an alcohol, after which the reaction product obtained reacts in situ with an alkanolamine and a polyesteramide is subsequently obtained through polycondensation.

20 Examples of suitable alcohols are (C₁-C₁₀) alcohols.

Preferably, methanol or ethanol is used.

In addition to hydroxyalkylamide groups, the polymer may also contain carboxyl groups, in amounts of between 0.01 and 2.0 mg equivalent/gram of polymer. The number of carboxylic acids present in the polymer can be controlled via the anhydride/alkanolamine ratio and via the degree of conversion. If an alkanolamine excess is used and the polycondensation reaction is (almost) complete, less than 0.2 mg equivalent acid/gram of polymer is usually present. If carboxyl groups are present, they may in a subsequent step react with compounds containing one or more groups that can react with carboxylic acid, such as for example epoxy groups or β-hydroxyalkylamide

groups. The amount of carboxylic acid is preferably as low as possible, for example between 0.01 and 0.2 mg equivalent/gram of polymer.

The degree of branching and the
5 functionality of the polymer are dependent on the starting materials and the molecular weight of the polymer. A molecular weight higher than 2000 and the use of di- and/or trialkanolamines generally lead to highly branched structures with a functionality of \geq
10 10.

Due to the presence in amounts of less than 10% by weight (of the total amount of anhydrides) of bis- and dianhydrides instead of the anhydrides according to formula (IX) it is possible that the
15 polymer does not comprise only products according to formulas (III) - (V).

The invention also relates to entirely or partly modified polymers.

The modification can for example take place
20 via a reaction between the polymer according to any one of formulas (III), (IV) or (V) with a monomer, oligomer or polymer containing reactive groups that can react with the hydroxyalkylamide.

Examples of suitable reactive groups
25 include carboxyl groups, carboxylic esters, carboxylic anhydrides, epoxy groups, alkoxysilane groups, isocyanate groups, acid chloride groups, epoxychlorohydrine groups, amine groups, phenolic groups, methylolated amidegroups and combinations
30 hereof.

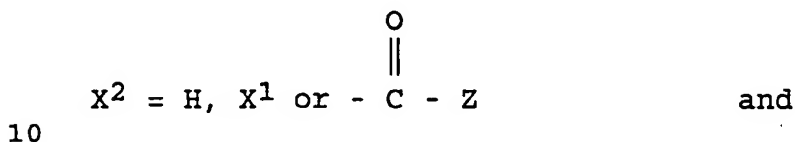
Preferably the monomer, oligomer or polymer contains only one group that can react with hydroxyalkylamide, as a result of which no crosslinking takes place during the modification.

35 The polymer according to formula (III),

(IV) or (V) has preferably been modified with a compound containing a carboxylic acid group.

A modified polymer can for example be represented by one of the formulas (III), (IV) or (V)

5 in which



in which $-\overset{\text{O}}{\parallel}\text{C}-Z$ is derived from a monomeric, oligomeric or polymeric monofunctional carboxylic acid.

15 Suitable carboxylic acids are, for example, saturated aliphatic (C_1 - C_{26}) acids, unsaturated (C_1 - C_{20}) fatty acids, aromatic acids and α,β -unsaturated acids.

Examples of suitable α,β -unsaturated acids
20 are (meth)acrylic acid, crotonic acid and monoesters or monoamides of itaconic acid, maleic acid, 12-hydroxystearic acid, polyether carboxylic acid, and fumaric acid.

Suitable saturated aliphatic acids are for
25 example acetic acid, propionic acid, butyric acid, 2-ethyl hexanoic acid, laurylic acid and stearic acid.

Suitable aromatic acid are for example benzoic acid and tertiary butyl benzoic acid.

Z can be chosen from, for example, a
30 saturated or unsaturated (C_1 - C_{40}) alkyl or aromatic group, a polymer or an oligomer. Examples of suitable polymers are polyesters, polyethers and poly(caprolactones).

Z can be substituted with for example ester
35 groups, ether groups, amide groups and alcohol groups.

The modified polymer may consist of the

same or different Z groups.

The branched polymer according to the invention can also react with a diisocyanate, after which the isocyanate-functional polymer obtained reacts
5 with a compound capable of reacting with isocyanates. As the diisocyanate use is preferably made of a compound containing two or more isocyanate groups with different reactivities. This is preferably an aliphatic diisocyanate with one sterically more accessible
10 isocyanate group bound to a primary carbon atom and one sterically less accessible isocyanate group bound to a tertiary carbon atom.

Examples of suitable diisocyanates are 1,4-diisocyanato-4-methyl-pentane, 1,5-diisocyanato-5-methylhexane, 3(4)-isocyanatomethyl-1-methylcyclohexylisocyanate, 1,6-diisocyanato-6-methyl-heptane, 1,5-diisocyanato-2,2,5-trimethylhexane and 1,7-diisocyanato-3,7-dimethyloctane, and 1-isocyanato-1-methyl-4-(4-isocyanatobut-2-yl)-cyclohexane, 1-
20 isocyanato-1,2,2-trimethyl-3-(2-isocyanato-ethyl)-cyclopentane, 1-isocyanato-1,4-dimethyl-4-isocyanatomethyl-cyclohexane, 1-isocyanato-1,3-dimethyl-3-isocyanatomethyl-cyclohexane, 1-isocyanato-1-n-butyl-3-(4-isocyanatobut-1-yl)-cyclopentane and 1-
25 isocyanato-1,2-dimethyl-3-ethyl-3-isocyanatomethyl-cyclopentane, respectively.

The preferred isocyanates are 3(4)-isocyanato-methyl-1-methylcyclohexylisocyanate (IMCI) and isophorone diisocyanate.

30 Monomers, oligomers and polymers can all be used as the compounds that can react with isocyanate groups. Such compounds contain reactive groups that can form a chemical bond with isocyanate groups.

Examples of suitable reactive groups are
35 alcohols and amine groups.

Examples of suitable compounds are hydroxyethyl(meth)acrylate, hydroxy(C₂-C₁₂)alkyl vinyl ether, 4-hydroxybutyl(meth)acrylate, aminopropyl vinyl ethers, aminoalkyl vinyl ether, aminopropyl-
5 tri(m)ethoxysilane and aminoalkyltrialkoxysilane.

Preferably the diisocyanate, for example IMCI, is combined with a selective catalyst, as a result of which no chain lengthening or crosslinking will take place.

10 As the catalyst use can be made of an ionogenic metal complex based on a metallic element from any one of groups III, IV or VII of the Periodic System with exchangeable counterions. Examples of suitable catalysts are titanium (IV) butoxide,
15 zirconium (IV) acetylacetonate, zirconium (IV) butoxide, tin (IV) acetate, manganese (III) acetylacetonate, titanium (IV) isopropoxide, zirconium (IV) 2-ethylhexanoate and tin (IV) chloride.

The modified and the unmodified polymers
20 can be very widely used in technically different fields, both in thermosetting and in thermoplastic applications. Examples are powder-paint compositions, coating systems based on water or solvent, can- or coil-coating systems, radiation-curable coating
25 compositions, alkyd resins for coatings, unsaturated resins for construction purposes (for example putties, sealants, castings, compounds and molding compounds), inks, toners, film formers for glass fibre sizings, adhesives, hot melts and in rubber compositions.

30 Unmodified or partly modified polymers according to the invention will generally be used in powder-paint systems, in can- or coil-coating systems or in solvent-based coating systems.

If the modification has been realized with

the aid of for example fatty acids, the polymers according to the invention can be used as airdrying systems.

A modification with radically curable compounds offers possibilities in the technical fields of radiation-curable coatings and construction resins.

Considering the many possibilities of modification of the polymer according to the invention, modification can be directed at any of a wide range of technical applications.

The polymers according to the invention can be used in thermosetting powder-paint compositions. Preferably use is made of the polymers containing β -hydroxyalkylamide groups.

Thermosetting powder paints have a better resistance to chemicals than thermoplastic powder paints. As a result of this, intensive efforts have for a long time been made to develop crosslinkers and polymers for thermosetting powder coatings. Attempts are still being made to find binder compositions for thermosetting powder paints with a good flow behaviour, good storage stability and a good reactivity. A thermosetting powder-paint binder composition generally contains more than 50 wt.% polymer and less than 50 wt.% crosslinker.

The polymer according to the invention can be used in a powder-paint composition as a polymer and as a crosslinker.

The glass transition temperature (T_g) of the polymer according to the invention lies between 0°C and 150°C, preferably between 50°C and 110°C, depending on the selected starting materials and the molecular weight.

Preferably a compound according to any one

of formulas (I), (II), (III), (IV) or (V) is used in powder-paint compositions. It is also possible to use a polymer in which up to for example 50 wt.%, preferably less than 30 wt.%, of the hydroxyalkylamide groups are
5 modified.

A coating that ultimately obtained with a powder paint must meet many varying requirements. Various systems are known. Some systems release volatile components during the curing. These systems
10 present the drawback that they form coatings with bubbles and/or that undesirable emissions are released. As far as the latter is concerned, the volatile component, if organic in origin, may cause undesirable environmental or health problems. It has moreover been
15 found that all the desired properties of the powder paint or powder coating are not always realized.

Systems comprising hydroxyalkylamide crosslinkers, such as for example according to EP-A-322834, contain bubbles above a layer thickness limit
20 of about 100 μm as a result of the reaction water released.

In other systems use is made of polyesters and the usual crosslinkers containing an epoxy group. No volatile components are generally released from
25 these systems. However, the use of bisphenol-A-epoxy resins in the so-called hybrid systems results in coatings that exhibit a relatively great extent of yellowing and powdering when exposed to UV light, while the frequently used triglycidylisocyanurate (TGIC)
30 crosslinker is toxicologically suspect.

It has been found that use of the polymer according to the invention as the crosslinker in binder compositions for powder paints results in a combination of highly desirable properties such as for instance

good flow behaviour and good resistance to chemicals, desired gloss without bubble formation at the surface up to and including layer thicknesses of at least 120 μm , a high resistance to scratching, good mechanical
5 properties, good powder stability, good weather resistance and good colour stability of the powder coating.

It is surprising that use of the highly functional crosslinkers according to the invention
10 leads to good flow behaviour, because generally a crosslinker having a functionality higher than, for example, 6 results in reduced flow behaviour.

Depending on the final application desired, the crosslinker according to the invention described
15 above can also be used in combination with another crosslinker, such as for example triglycidyl isocyanurate (TGIC), polybisphenol-A-epoxides such as, for instance, the various EpikoteTM grades, compounds containing (blocked) isocyanate groups, such as for
20 example the caprolactam-blocked isophorone diisocyanate trimer, crosslinkers containing β -hydroxyalkylamide

groups such as for example Primid XL 522TM (Rohm and Haas) and/or polyfunctional oxazolines. The weight ratio between the crosslinkers can be selected
25 depending on the final application.

The crosslinker according to the invention is preferably combined with a crosslinker comprising at least one linear or branched aliphatic chain with 5-26 carbon atoms and having an epoxy functionality of more
30 than 1, with the proviso that the epoxy groups are carried on the at least one aliphatic chain. These crosslinkers are described in EP-A-600546 and include, for example, epoxidized oils in which the oil is linseed oil, soybean oil, safflower oil, oiticica oil,

carraway seed oil, rapeseed oil, castor oil, dehydrated
castor oil, cottonseed oil, wood oil, vernonia oil (a
natural oil), sunflower oil, peanut oil, olive oil,
soybean oil, maize oil, fish oil such as, for instance,
5 herring or sardine oil, and non-cyclic terpene oils.

The epoxidized oil is preferably epoxidized
soybean oil and/or epoxidized linseed oil.

As the crosslinker, a powder-paint-binder
composition may contain the polymer according to the
10 invention and as the polymer a polymer containing
carboxyl groups or containing anhydride groups.

A polyester, a polyacrylate, a polyether
(such for example a polyether based on bisphenol or a
phenol-aldehyde novolak), a polyurethane, a
15 polycarbonate, a trifluoroethylene copolymer or a
pentafluoropropylene copolymer, a polybutadiene, a
polystyrene or a styrene maleic anhydride copolymer can
for example be chosen as the polymer.

Generally, polymers having an acid value
20 higher than 40 mg KOH/gram resins are applied because a
relatively high acid value results in better reactivity
with the polymer according to the invention.

The molecular weight (M_n) of this polymer
is usually higher than 800, but preferably higher than
25 1500. The polymer must flow well at temperatures
between 100°C and 200°C and therefore has a molecular
weight (M_n) that is lower than approximately 10,000,
preferably lower than approximately 7000.

This polymer generally has a viscosity at
30 158°C that is lower than 8000 dPas. The viscosity will
usually be higher than 100 dPas. The viscosity can
advantageously vary from approximately 300 to
approximately 5000 dPas. The viscosity used here was
measured according to the Emila method described by

Misev in Powder Coatings; Chemistry and Technology, pages 287-288 (1991).

The Tg of this polymer is generally higher than approximately 20°C, preferably higher than 30°C, and may be higher than 40°C. The polymer's Tg is usually lower than 120°C because otherwise the binder composition may become somewhat difficult to prepare. As already indicated above, the choice of the polymer's Tg can be based on the Tg recommended for the binder composition.

If use is made of polymers having only terminal groups that can react with a hydroxyalkylamide functionality, the polymer has an average functionality (capable of reacting with the hydroxyalkylamide groups) of more than 1.6, preferably more than 2. The polymer generally has an average functionality of less than 10, preferably less than approximately 6. If use is made of polymers - such as polyacrylates - with appended functional groups, the average functionality will be higher than approximately 1.6, and preferably higher than 2. Such a polymer generally has an average functionality of less than 8, preferably less than 4.

Most preferable of the suitable polymers are polyesters and polyacrylates.

With the polymers described various properties can be obtained in the binder and in the powder coating itself. Polyacrylates are highly resistant to yellowing and to weather influences. The polyacrylates that can be used as the polymer may be based on (meth)acrylic acid, methyl(meth)acrylate, ethyl(meth)acrylate, butyl(meth)acrylate, propyl(meth)acrylate, 2-ethylhexyl(meth)acrylate, cyclohexyl(meth)acrylate, decyl(meth)acrylate, isodecyl(meth)acrylate, benzyl(meth)acrylate and

hydroxyalkyl(meth)acrylates such as hydroxyethyl and hydroxypropyl(meth)acrylate and/or glycidyl esters or glycidyl ethers of alkyl(meth)acrylates.

The polyacrylates can be obtained via known processes. In these processes use can be made of comonomers such as for instance styrene, maleic acid or maleic anhydride and of small amounts of ethylene, propylene and acrylonitrile. Other vinyl or alkyl monomers, such as octene, triallyl isocyanurate and diallyl phthalate, can be added in small amounts.

A polyacrylate containing acid groups is generally obtained through copolymerization of the desired amount of acid, such as for example (meth)acrylic acid, maleic acid or fumaric acid.

The polyacrylate's viscosity usually lies between 100 and 8000 dPas (measured at 158°C; Emila).

Polyacrylates are described in the patents US-A-3,752,870, US-A-3,787,340 and US-A-3,758,334 and in the British patent 1,333,361, and what is disclosed in said patents is included herein by means of this reference.

The polyurethanes that can be used as the polymer that can react with β -hydroxyalkylamide groups include for example also the polyurethanes terminated with an acid group and a (blocked) isocyanate group.

Polyesters are usually based on the residues of aliphatic polyalcohols and polycarboxylic acids.

The polycarboxylic acids are generally chosen from the group consisting of aromatic and cycloaliphatic polycarboxylic acids because these acids usually have a Tg-raising effect on the polyester. In particular, use is made of dibasic acids. Examples of polycarboxylic acids are isophthalic acid, terephthalic

acid, hexahydroterephthalic acid, 2,6-naphthalene dicarboxylic acid and 4,4'-oxybisbenzoic acid and, subject to availability, their anhydrides, acid chlorides or lower alkyl esters, such as for example the dimethyl ester of naphthalene dicarboxylic acid. Although not required, the carboxylic acid component generally contains at least approximately 50 mol.%, preferably at least approximately 70 mol.%, isophthalic acid and/or terephthalic acid.

10 Other suitable aromatic cycloaliphatic and/or acyclic polycarboxylic acids are for example 3,6-dichlorophthalic acid, tetrachlorophthalic acid, tetrahydrophthalic acid, hexahydroterephthalic acid, hexachloroendomethylene tetrahydrophthalic acid, 15 phthalic acid, azelaic acid, sebacic acid, decanedicarboxylic acid, adipic acid, succinic acid, trimellitic acid and maleic acid. These different carboxylic acids can be used in amounts of at most 50 mol.% of the total amount of carboxylic acids. These 20 acids can be used as such or, subject to availability, in the form of their anhydrides, acid chlorides or lower alkyl esters.

 Hydroxycarboxylic acids and/or optionally lactones can also be used, for example 12- 25 hydroxystearic acid, hydroxypivalic acid and ϵ -caprolactone. If so desired, monocarboxylic acids, such as benzoic acid, tert.-butylbenzoic acid, hexahydrobenzoic acid and saturated aliphatic monocarboxylic acids can be used in smaller amounts.

30 The polyalcohols, in particular diols, that can be caused to react with the carboxylic acids to obtain the polyester include aliphatic diols such as for example ethylene glycol, propane-1,2-diol, propane-1,3-diol, butane-1,2-diol, butane-1,4-diol, butane-1,3-

diol, 2,2-dimethylpropanediol-1,3 (= neopentyl glycol), hexane-2,5-diol, hexane-1,6-diol, 2,2-bis-(4-hydroxycyclohexyl)-propane (hydrogenated bisphenol-A), 1,4-dimethylolcyclohexane, diethylene glycol, dipropylene glycol and 2,2-bis[4-(2-hydroxyethoxy)-phenyl]propane and the hydroxypivalic ester of neopentyl glycol.

Small amounts, such as less than approximately 4 wt.%, but preferably less than 2 wt.%, of trifunctional alcohols or acids can be used to obtain branched polyesters. Examples of suitable polyols and polyacids are glycerol, hexanetriol, trimethylolethane, trimethylolpropane, tris-(2-hydroxyethyl)-isocyanurate and trimellitic acid.

Tetrafunctional monomers are usually not preferred, because they can cause excessive branching or gelling, although it is possible to use them in very small amounts. Examples of suitable polyfunctional alcohols and acids are sorbitol, pentaerythritol and pyromellitic acid. Trifunctional monomers are however preferred for synthesizing branched polyesters.

The coating properties can be influenced via for example the choice of diol. If for example good weather resistance is required, the alcohol component preferably contains at least 70 mol.% neopentyl glycol, 1,4-dimethylolhexane and/or hydrogenated bisphenol-A. Caprolactone and hydropivalic acid can also be used if good weather resistance is required.

The polyesters are prepared via the usual processes, through esterification or transesterification, optionally in the presence of the usual esterification catalysts such as for example dibutyl tin oxide or tetrabutyl titanate. The preparation conditions and the COOH/OH ratio can be chosen so that end products having an acid number or hydroxyl value that lies within the desired range of values are

obtained.

A carboxylic-acid-functional polyester is preferably prepared in a series of steps. In the last step thereof an aromatic or, preferably, aliphatic acid is esterified so that an acid-functional polyester is obtained. As known to a person skilled in the art, terephthalic acid is in a first step caused to react in the presence of excess diol. Such reactions result in a substantially hydroxyl-functional polyester. In a second or subsequent step an acid-functional polyester is obtained by causing further acid to react with the product of the first step. Further acids are for example isophthalic acid, adipic acid, succinic anhydride, 1,4-cyclohexanedicarboxylic acid and trimellitic anhydride.

Preferably trimellitic anhydride is used at a temperature of 170-200°C, because then a polyester with a relatively large number of trimellitic acid terminal groups is obtained, as a result of which the reactivity of the binder system is increased and better coating properties are obtained.

The polyester may be a crystalline polyester, but amorphous polyesters are preferred. Mixtures of crystalline and amorphous polyesters can also be used. Amorphous polyesters have a viscosity that generally lies within a range from 100 to 8000 dPas (measured at 158°C, Emila). Crystalline polyesters usually have a lower viscosity in the range from approximately 2 to approximately 200 dPas.

If the polyester contains groups that can react with carboxylic acid, the polyester's acid number is chosen so that the desired amount of crosslinker can be used. The acid number is preferably higher than 10 and more preferably higher than 40.

The polyester's Tg is chosen so that the Tg of the polyester-crosslinker mixture lies between for example 30°C and 80°C, as a result of which powder paints or binders prepared from them are physically
5 stable at room temperature. Combinations of polyester and crosslinker having a lower Tg can optionally be used in the preparation of a powder coating composition. To retain the powder stability, such powders are however stored in cooled condition.

10 The selection of the polymer : crosslinker weight ratio depends on the desired final application and this ratio will generally be between 60:40 and 90:10, preferably between 75:25 and 85:15.

If the polymer according to the invention
15 is used as a resin in powder-paint compositions, compounds containing two or more functional groups that can react with β -hydroxyamide groups can be used as the crosslinker. Examples of such groups are anhydrides, carboxylic acids, carboxylic esters, epoxides,
20 isocyanates and alkoxysilanes. Preferably anhydride groups, carboxylic acids and blocked isocyanates are

used. Examples are adipic acid, decanedicarboxylic acid, trimellitic anhydride, phthalic acid or phthalic anhydride, tetrahydrophthalic acid or tetrahydro-
25 phthalic anhydride, hexahydrophthalic acid or hexahydrophthalic anhydride and IPDI-trimer or HDI-trimer, optionally blocked with caprolactam or triazole.

The preparation of thermosetting powder
30 coatings in general and the chemical reactions for curing powder paints to form cured coatings are described by Misev in Powder Coatings, Chemistry and Technology (1991, John Wiley) on pp. 42-54, pp. 148 and 224-226. A thermosetting binder composition is

generally defined as the resinous part of the powder paint consisting of polymer and crosslinker.

If so desired, the usual additives can be used in the binder composition and in the powder-paint system according to the invention, such as for example pigments, fillers, degassing agents, flow agents and stabilizers. Suitable pigments are for example inorganic pigments, such as for example titanium dioxide, zinc sulphide, iron oxide and chromium oxide, and also organic pigments such as for example azo compounds. Suitable fillers are for example metal oxides, silicates, carbonates and sulphates.

Primary and/or secondary antioxidants, UV stabilizers such as quinones, (sterically hindered) phenolic compounds, phosphonites, phosphites, thioethers and HALS compounds (hindered amine light stabilizers) can for example be used as stabilizers.

Examples of degassing agents are benzoin and cyclohexane dimethanol bisbenzoate. The flow agents include for example polyalkylacrylates, fluorohydrocarbons and silicone fluids. Other suitable additives are for example additives for improving tribocharging, such as sterically hindered tertiary amines that are described in EP-B-371528.

Powder paints according to the invention can be applied in the usual manner, for example by electrostatically spraying the powder onto an earthed substrate and curing the coating by exposing it to heat at a suitable temperature for a sufficient length of time. The applied powder can for example be heated in a gas oven, an electric oven or with the aid of infrared radiation.

Thermosetting coatings of powder-paint (coating) compositions intended for industrial applications are described further in a general sense

in Powder Coatings, Chemistry and Technology, Misev, pages 141-173 (1991).

Compositions according to the present invention can be used in powder paints for use on, for example, metal, wooden and plastic substrates. Examples are industrial coatings, coatings for machines and tools, household applications and parts of buildings. The coatings are also suitable for use in the automotive industry for coating parts and accessories.

DE-A-19703952 discloses a copolyester containing β -hydroxyalkylamide groups as endgroups. The polyester polymer backbone does not comprise amide groups. The copolyester is prepared in a three step process by mixing a hydroxy polyester with a polycarboxylic acid dialkyl ester to form an alkylester group containing copolyester followed by reaction with an aminoalcohol. In contrast, the polymer according to the invention is a polyesteramide having amide and ester groups along the backbone in addition to the β -hydroxyalkylamide endgroups. This polyesteramide results in improved mechanical coating properties by more extensive hydrogen bridge formation, improved crosslinkdensity and improved hydrolysis resistance.

The invention will be elucidated with reference to the following, non-limiting examples.

Example I

Preparation of a highly branched polymer comprising units of phthalic anhydride and diisopropanolamine

384 g of phthalic anhydride and 415 g of diisopropanolamine were introduced into a double-walled glass reactor, which could be heated by means of thermal oil, fitted with a mechanical stirrer, a distillation head and nitrogen and vacuum connections.

The reaction mixture was gradually heated, with stirring, to approx. 70°C and then more slowly to 170°C. A vacuum was created during the heating. The pressure in the reactor was adjusted to the release of reaction water, so that this could be removed from the reactor through distillation. After a total reaction time of 6 hours the viscous polymer contained less than 0.1 meq/g carboxylic acid (titrimetrically determined) and no more water could be removed through distillation. After cooling the polymer was obtained as a very pale yellow glassy mass. The concentration of hydroxyl groups was titrimetrically found to be 5.4 meq/g. The number average molecular mass was determined with the aid of GPC (universal calibration) and was 1500 g/mol; the weight average molecular mass was 7700 g/mol.

Example II

20 Preparation of a highly branched polymer comprising units of phthalic anhydride and diisopropanolamine

232 g of phthalic anhydride and 270 g of diisopropanolamine were introduced into a double-walled glass reactor, which could be heated by means of thermal oil, fitted with a mechanical stirrer, a distillation head and nitrogen and vacuum connections. The reaction mixture was gradually heated, with stirring, to approx. 70°C and then more slowly to 170°C. A vacuum was created during the heating. The pressure in the reactor was adjusted to the release of reaction water, so that this could be removed from the reactor through distillation. After a total reaction time of 5 hours the viscous polymer contained less than 0.2 meq/g of carboxylic acid (titrimetrically

determined) and no more water could be removed through distillation. After cooling the polymer was obtained as a pale yellow glassy mass. The concentration of hydroxyl groups was titrimetrically found to be 5.8 meq/g. The number average molecular mass was determined with the aid of GPC (universal calibration) and was 1100 g/mol; the weight average molecular mass 4900 g/mol.

10 Example III

Preparation of a highly branched polymer comprising units of hexahydrophthalic anhydride and diisopropanolamine

398 g of hexahydrophthalic anhydride and 408 g of diisopropanolamine were introduced into a double-walled glass reactor, which could be heated by means of thermal oil, fitted with a mechanical stirrer, a distillation head and nitrogen and vacuum connections. The reaction mixture was gradually heated, with stirring, to approx. 70°C and then more slowly to 160°C. A vacuum was created during the heating. The

pressure in the reactor was adjusted to the release of reaction water, so that this could be removed from the reactor by means of distillation. After a total reaction time of 3.5 hours the viscous polymer contained less than 0.2 meq/g of carboxylic acid (titrimetrically determined) and no more water could be removed through distillation. After cooling the polymer was obtained as an almost colourless glassy mass. The concentration of hydroxyl groups was titrimetrically found to be 5.2 meq/g. The number average molecular mass was determined with the aid of GPC (universal calibration) and was 1550 g/mol; the weight average molecular mass 7000 g/mol.

Example IVPreparation of a highly branched polymer comprising units of hexahydrophthalic anhydride and5 diisopropanolamine

378 g of hexahydrophthalic anhydride and 436 g of diisopropanolamine were introduced into a double-walled glass reactor, which could be heated by means of thermal oil, fitted with a mechanical stirrer, a distillation head and nitrogen and vacuum connections. The reaction mixture was gradually heated, with stirring, to approx. 70°C and then more slowly to 160°C. A vacuum was created during the heating. The pressure in the reactor was adjusted to the release of reaction water, so that this could be removed from the reactor through distillation. After a total reaction time of 5 hours the viscous polymer contained less than 0.1 meq/g carboxylic acid (titrimetrically determined) and no more water could be removed through distillation. After cooling the polymer was obtained as an almost colourless glassy mass. The concentration of hydroxyl groups was titrimetrically found to be 6.1 meq/g. The number average molecular mass was determined with the aid of GPC (universal calibration) and was 1010 g/mol; the weight average molecular mass 4600 g/mol.

Examples V-VIIIPowder-paint compositions comprising a polymer30 according to any one of Examples I-IV

Powder-paint compositions according to Table 1 were prepared by mixing and extrusion (PRISM extruder, 120°C). The polyesters (Uralac 5040TM and Uralac 5261TM from DSM Resins) comprise units of

terephthalic acid, adipic acid, neopentyl glycol and trimellitic anhydride.

The compositions were in the usual manner ground, sieved and electrostatically sprayed (Corona)
5 onto aluminium and steel test panels. After a cure cycle of 10 minutes at 200°C or 15 minutes at 180°C in a circulation oven, the panels were tested to determine their appearance (visually), flexibility (penetration in mm according to Erichsen ISO 1520/DIN 53156),
10 reverse impact resistance (ASTM-2794/69 in inch-pound), acetone resistance (acetone double rubs), adhesion (cross hatch adhesion test) and hardness (König, seconds). The test results are shown in Table 1.

These examples show that the polymers
15 according to the invention result in coatings having good to very good chemical, mechanical and optical properties and a high blister limit (visually).

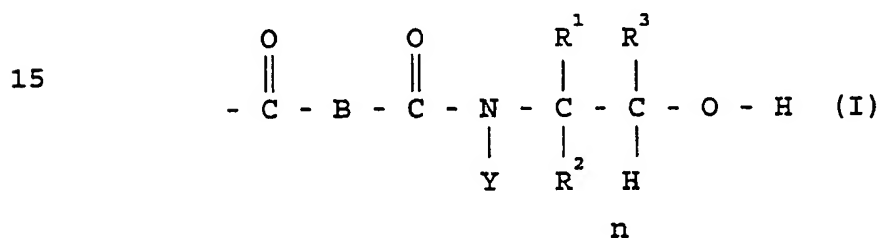
TABLE 1: Compositions and coating properties

Composition	A	B	C	D	E	F
Polyester resin: Uralac P5261 TM Uralac P5040 TM	155 g	156 g	162 g	159 g	160 g	164 g
Crosslinker according to: Example I Example II Example III Example IV	45 g	44 g	38 g	43 g	42 g	36 g
Additives: TiO ₂ 2160 Benzoin BYK 361	100 g 3.0 g 1.5 g	100 g 3.0 g 1.5 g	100 g 3.0 g 1.5 g	100 g 3.0 g 1.5 g	100 g 3.0 g 1.5 g	100 g 3.0 g 1.5 g
cure cycle	10' 200°C	10' 200°C	10' 200°C	15' 180°C	15' 180°C	15' 180°C
Hardness Impact resistance ¹⁾ ESP ²⁾ Adhesion ³⁾ Gel time ⁴⁾ Flow behaviour Blister limit	235 s > 160ip > 8 mm Gt0 n.d.5) OK 120 μ	225 s 160 ip > 8 mm Gt0 151 OK 120 μ	215 s > 160 ip > 8 mm Gt0 80 OK 130 μ	215 s > 160 ip > 8 mm Gt0 116 OK 140 μ	210 s > 160 ip > 8 mm Gt0 85 OK 140 μ	195 s > 160 ip > 8 mm Gt0 100 OK 140 μ

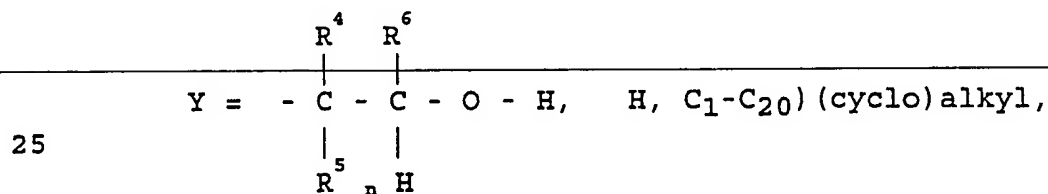
- 1) reverse impact test; on steel ASTM-2794/69.
The impact resistance is usually given as
inch x pound. If no cracks are visible in the
5 coating at 160 i.p., 160 i.p. is quoted as
the result. An impact resistance of 160 i.p.
stands for 1.84 m.kg.
 - 2) Erichsen Slow Penetration; ISO 1520/DIN 53156
 - 3) Cross-hatch adhesion; ISO 2409/DIN 5315
 - 10 4) DIN 55990; part B.
 - 5) not determined.
-

C L A I M S

- 5 1. Condensation polymer containing ester groups and at least one amide group in the backbone, having at least one hydroxyalkylamide end group and having a weight average molecular mass of ≥ 800 g/mol.
- 10 2. Polymer according to Claim 1, characterized in that the polymer contains at least two groups according to formula (I):



20 in which

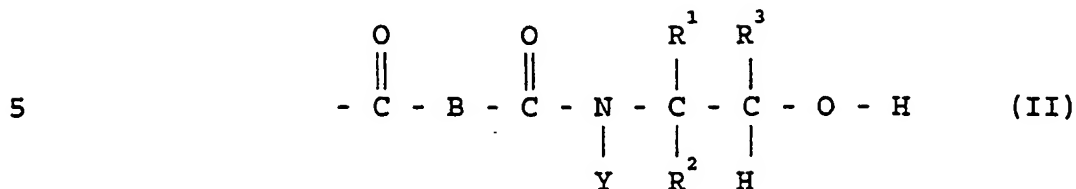


or (C₆-C₁₀) aryl,

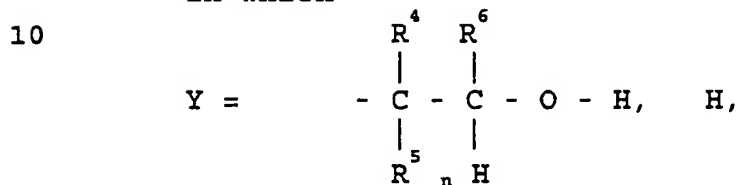
B = (C₂-C₂₄), optionally substituted, aryl or
 30 (cyclo)alkyl aliphatic diradical,
 R¹, R², R³, R⁴, R⁵ and R⁶ may, independently of one another, be the same or different, H, (C₆-C₁₀) aryl or (C₁-C₈) (cyclo)alkyl radical and
 n = 1-4.

- 35 3. Polymer according to any one of Claims 1-2, characterized in that the polymer comprises at

least two groups according to formula (II):



in which



(C₁-C₂₄) (cyclo)alkyl or (C₆-C₁₀) aryl,

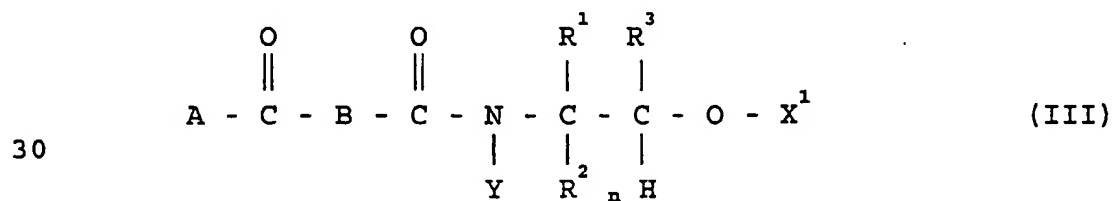
B = (C₂-C₂₀), optionally substituted, aryl or

(cyclo)alkyl aliphatic diradical, and

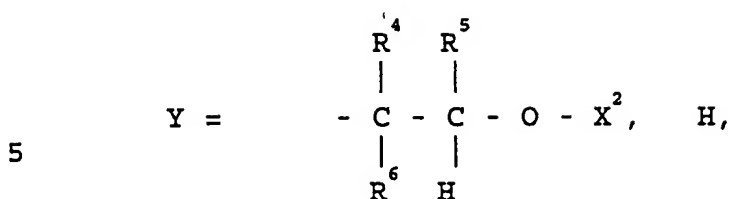
20 R¹, R², R³, R⁴, R⁵ and R⁶ may, independently of one another, be the same or different, H, (C₆-C₁₀) aryl or (C₁-C₈) (cyclo)alkyl radical.

4. Condensation polymer according to any one of

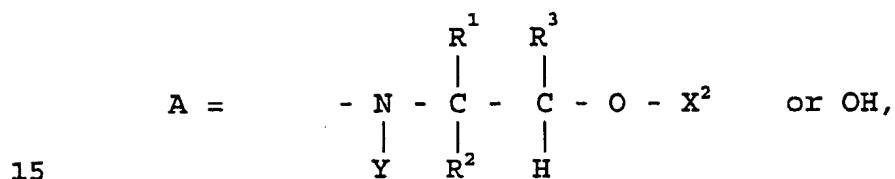
25 Claims 1-3, characterized in that the polymer is a polymer according to formula (III):



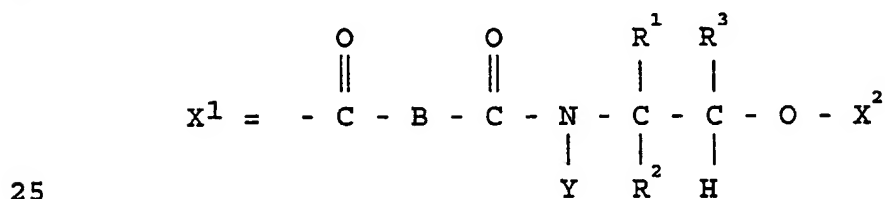
in which:



10 (C₁-C₂₀) (cyclo)alkyl or (C₆-C₁₀)aryl,



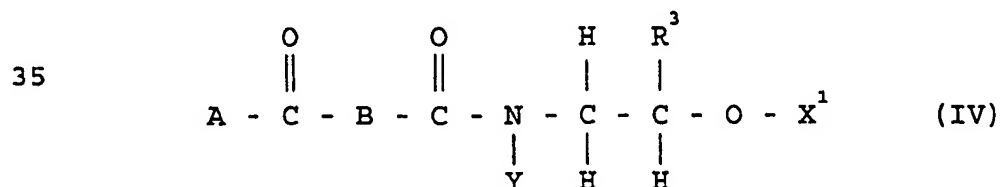
20 B = (C₂-C₂₄), optionally substituted, aryl or (cyclo)alkyl aliphatic diradical,



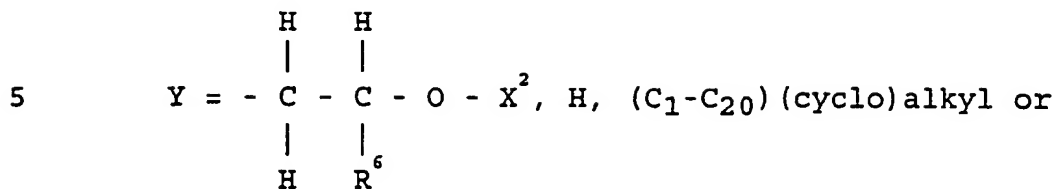
X² = H or X¹ and

R¹, R², R³, R⁴, R⁵ and R⁶ may be H, (C₆-C₁₀) aryl or (C₁-C₈) (cyclo)alkyl radical or CH₂-OX².

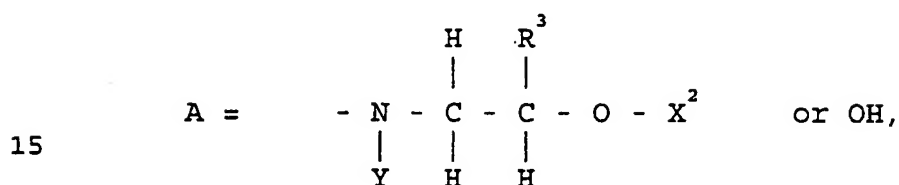
30 5. Condensation polymer according to any one of Claims 1-4, characterized in that the polymer is represented by formula (IV):



in which:

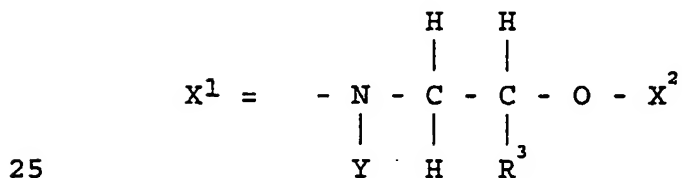


10 $(\text{C}_6\text{-C}_{10})\text{aryl}.$



$B = (\text{C}_2\text{-C}_{24}),$ optionally substituted, aryl or
(cyclo)alkyl aliphatic diradical,

20



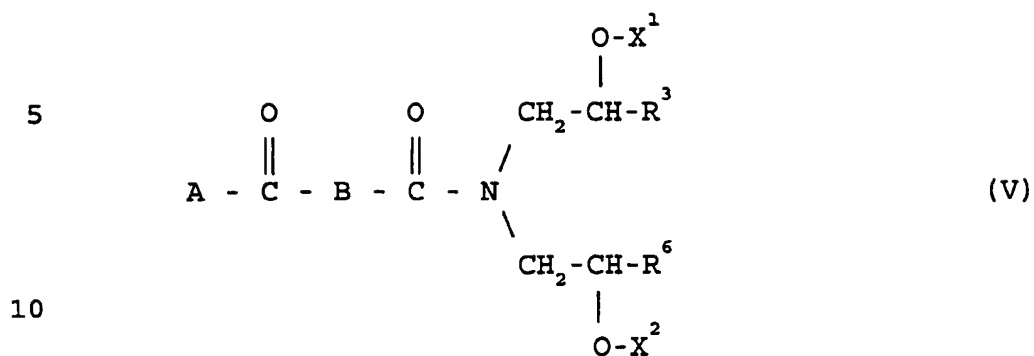
$X^2 = \text{H or } X^1,$

$R^3 = \text{H or } (\text{C}_6\text{-C}_{10}) \text{ aryl or } (\text{C}_1\text{-C}_8)\text{alkyl radical},$

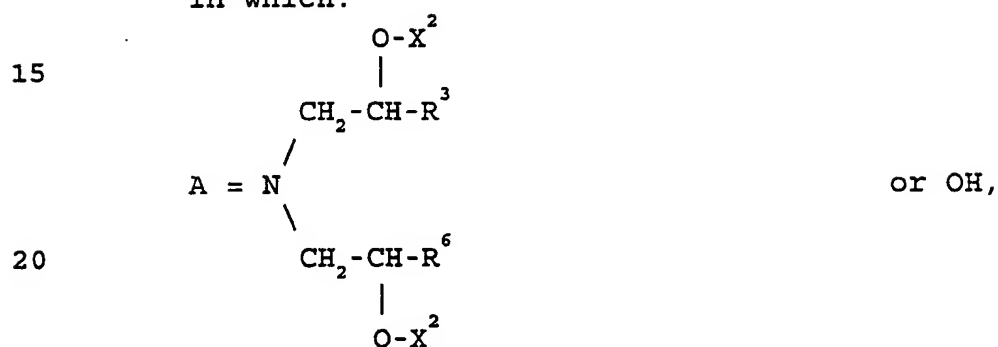
and

30 $R^6 = \text{H or } (\text{C}_6\text{-C}_{10}) \text{ aryl or } (\text{C}_1\text{-C}_8)\text{alkyl radical}.$

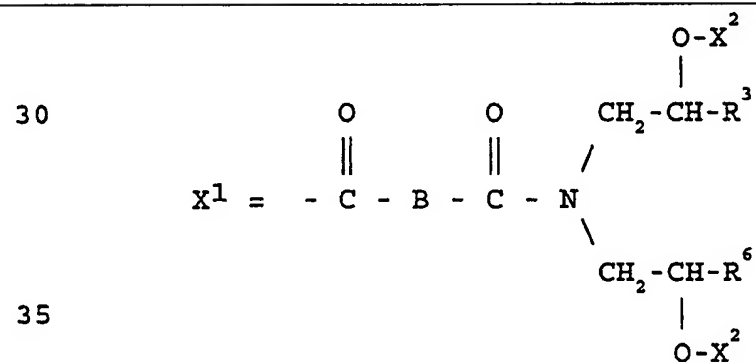
6. Condensation polymer according to any one of
Claims 1-5, characterized in that the polymer is a
polymer according to formula (V):



in which:



25 $\text{B} = (\text{C}_2 - \text{C}_{12})$, optionally substituted, aryl or (cyclo)alkyl aliphatic diradical,



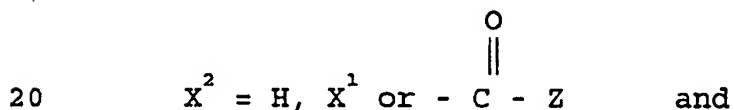
$\text{X}^2 = \text{H}$ or X^1 ,

$\text{R}^3 = \text{H}$ or $(\text{C}_6 - \text{C}_{10})$ aryl or $(\text{C}_1 - \text{C}_8)$ (cyclo)alkyl radical and

40 $\text{R}^6 = \text{H}$ or $(\text{C}_6 - \text{C}_{10})$ aryl or $(\text{C}_1 - \text{C}_8)$ (cyclo)alkyl radical.

7. Polymer according to any one of Claims 2-6,
characterized in that B is a (methyl-)1,2-
ethylene, (methyl-)1,2-ethylidene, 1,3-propylene,
(methyl-)1,2-cyclohexyl, (methyl-)1,2-phenylene,
5 1,3-phenylene, 1,4-phenylene, 2,3-norbornyl, 2,3-
norbornen-5-yl and/or (methyl-)1,2-cyclohex-4-enyl
radical.
8. Polymer according to any one of Claims 1-7,
characterized in that the weight average molecular
10 mass is between 800 and 50,000 g/mol, the number
average molecular mass is between 600 and 10,000
and the hydroxyalkylamide functionality is between
2 and 250.
9. Polymer according to any one of Claims 1-8
15 obtainable by polycondensation of a mono- and/or
bis- β -hydroxyalkylamide of a bivalent carboxylic
acid.
10. Process for the preparation of a polymer according
to any one of Claims 1-9, characterized in that a
20 cyclic anhydride reacts with an alkanolamine to
form a hydroxylalkylamide, after which the polymer
is obtained through polycondensation.
11. Process for the preparation of a polymer according
to any one of Claims 1-9, characterized in that an
25 alkanolamine reacts with a compound containing an
acid group and an activated acid group, after
which the polymer is obtained through
polycondensation.
12. Process for the preparation of a polymer according
30 to any one of Claims 1-9, characterized in that a
cyclic anhydride reacts with an alcohol, after
which the reaction product obtained reacts in situ
with an alkanolamine and the polymer is
subsequently obtained through polycondensation.

13. Process according to any one of Claims 10-12, characterized in that the alkanolamine is a β -alkyl-substituted β -hydroxyalkylamide.
14. Process according to any one of Claims 10-13, characterized in that the alkanolamine is
 5 (di)isopropanolamine, 1-(m)ethylisopropanolamine, (di)isobutanolamine, cyclohexylisopropanolamine, di- β -cyclohexanolamine and/or n-butylisopropanolamine.
- 10 15. Modified polymer that can be obtained through a reaction between a polymer according to any one of formulas (III), (IV) or (V) with a monomer, oligomer or polymer containing reactive groups that can react with hydroxyalkylamide.
- 15 16. Modified polymer according to any one of formulas (III), (IV) or (V) in which



25
$$\overset{\text{O}}{\parallel}$$

 in which C - Z is derived from a monomeric, oligomeric or polymeric monofunctional carboxylic acid.

17. Powder-paint binder composition containing a polymer according to any one of Claims 1-9 or 15-
 30 16 or a polymer obtained according to any one of Claims 10-14.
18. Binder composition according to Claim 17 which additionally contains a crosslinker comprising at least one linear or branched aliphatic chain with
 35 5-26 carbon atoms and having an epoxy functionality of more than 1, with the proviso

that the epoxy groups are carried on the at least one aliphatic chain.

19. Powder paint composition comprising a binder composition according to any one of Claims 17-18.
 - 5 20. Powder coating obtained by curing of a powder paint according to Claim 19.
 21. Entirely or partly coated substrate, characterized in that a powder coating according to Claim 20 is used as the coating.
-

INTERNATIONAL SEARCH REPORT

Int'l. Application No

PCT/NL 98/00546

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C08G69/44 C08G63/685

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US 3 709 858 A (ALBERS K ET AL) 9 January 1973 -----	

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

21 October 1998

Date of mailing of the international search report

28/10/1998

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Information on patent family members

PCT/98/00546

Form PCT/ISA/210 (patent family annex) (July 1992)

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